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LOW TEMPERATURE RECOVERY OF RADIATION DAMAGE IN VANADIUM

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ABSTRACT

Some system may emerge from the numerous anomalies in the low temperature recovery of irradiated V if we assume that Stage I (first recovery by interstitial migration) occurs below 4 K. Present supporting evidence includes: from both thermal neutron and fission neutron damage studies -- (1) continuous, nearly structureless recovery from 4 to 43 K independent of dose; (2) a non-linear, decreasing damage rate at 4 K; and (3) from 6 K electron irradiations near threshold energy there is derived a Frenkel pair resistivity which is so small as to suggest significant loss of defects during irradiation. The assumption of a very low-lying Stage I also helps to explain several other unusual aspects of the data, especially those relating to the sharp recovery peak at 47 K. This is clearly a single first-order process (with a measured activation energy of 0.13 eV), but it has a strong *positive* dependence of population percentage upon dose, opposite to Stage II impurity detrapping peaks in several fcc metals. Also, the peak has a strong negative dependence upon irradiation temperature when equal doses put in at 4 and 31 K are compared.

INTRODUCTION

Our earlier work[1] established the overall low temperature recovery behavior of radiation damage in V as follows: (1) Thermal neutron damage and fission-spectrum fast neutron damage, when doses yielding equal damage resistivities are compared, exhibit remarkable similarity despite mean primary recoil energies calculated to be 433 eV and 20 keV, respectively. (2) Continuous, nearly structureless recovery starts within 1 K° of a 3.8 K irradiation temperature[2] and extends to 43 K to recover 20% of the damage resistivity independent of dose. (3) A sharply peaked recovery process at 47 K is stationary in temperature with dose variations, but it

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(4) grows in percentage with increasing dose, nearly doubling when initial damage resistivity is raised from 4 to 16 $n\Omega cm$. (5) The continuous recovery continues beyond this peak, with assorted peak structure above 60 K, the details of which will not be treated here.

The only other known studies of the low temperature damage production and recovery in vanadium[3,4] dealt with samples of much lower purity and with high or very high doses of fast neutrons (5 to 80 times our highest resistivity change). Both showed more-or-less continuously rising early recovery rates peaking broadly in the 45 to 80 K range with subpeaks especially at about 50 and 70 K. Qualitatively, this agrees fairly well with our observations.

Compared to Fe and some other bcc metals which show great similarity to the well-established classical fcc model of defect behavior, V seems anomalous. Especially unusual is the first observed recovery process, which spans a full decade in temperature without significant structure (no Stage $I_{A,B,C}$ close-pair peaks) despite low doses and moderate displacement energies. Also unusual is the sense of the dose dependence of the first peak, *growing* with dose while remaining stationary in temperature.

The present work was carried out to examine in more detail the unusual behavior already noted and to determine the effect of irradiation temperature on the 47 K recovery peak.

EXPERIMENTAL

A 0.25-mm-diameter wire sample of V was mounted in a two-turn spiral groove in an anodized aluminum disk so as to be transverse to the magnetic field of a superconducting solenoid surrounding the irradiation capsule. The measuring techniques were as described previously[1] except that with a 1.6 kOe magnetic field the resistivity measurements could be made below the V 5.1 K superconducting transition. The description of sample preparation and purity given for our prior study[1] applies here as well. This sample's residual resistivity ratio, RRR, was $R_{300K}/R_{3.8K} = 380$.

Damage Production.--The damage rate data plotted in Fig. 1 is for thermal neutron damage at 4.9 K during which 40 $n\Omega cm$ damage resistivity was put into the V sample starting from an initial resistivity of 53 $n\Omega cm$.

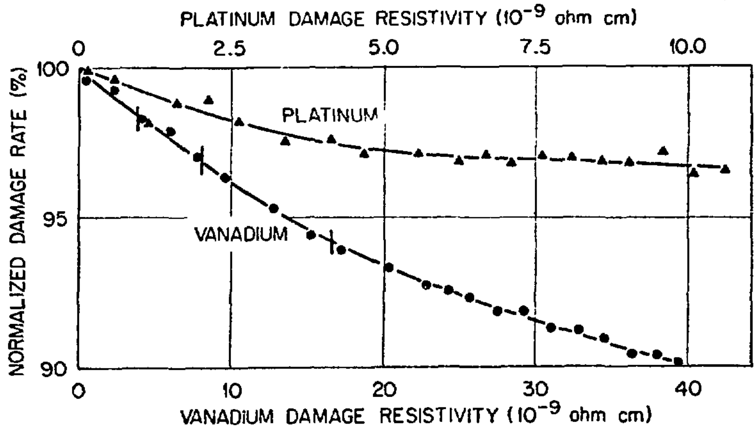

Fig. 1. Normalized damage rates in V and Pt at 4.9 K with thermal neutrons. $(d\rho/dc)/(d\rho/dc)_0$ c is dose.

Also shown are the data taken simultaneously for a Pt sample in the same capsule. Both samples show similar non-linear decreases in the rate, with a 10% drop in the V by the end of the run, and 3.5% in the Pt. Three tick marks on the V curve indicate the dose levels used for the low and high doses (4 and 16 nΩcm) of the earlier study[1] and the level (8 nΩcm) used in the measurement described next.

Dependence of 47 K Recovery on Damage Production Temperature.--Three complete damage and isochronal anneal runs were made, all with the same fluence. Each was preceded by a 20-min anneal at 420 K. In the first and third runs the irradiation temperature was 4.3 K, and for the intervening run it was 30.7 K. Isochronal recovery from 35 to 75 K was measured in each run, using the same temperature schedule in which $\Delta T/T = 4.0\%$. Results of the two 4.3 K runs were virtually identical, showing no influence from the slight accumulation of "old" defects not removed by the 420 K anneal. Recovery and recovery rate curves for runs 2 and 3 are plotted in Fig. 2. Damage resistivities remaining from either irradiation were virtually identical when both had been annealed to the same temperature, for any isochronal temperature in the range 35 to 43 K. The amount of recovery in the 47 K peak, after deducting the continuous background recovery, was half as much in the 30.7 K run as in the 4.3 K run (5% vs 10%, normalized to the damage resistivity remaining at 38.2 K).

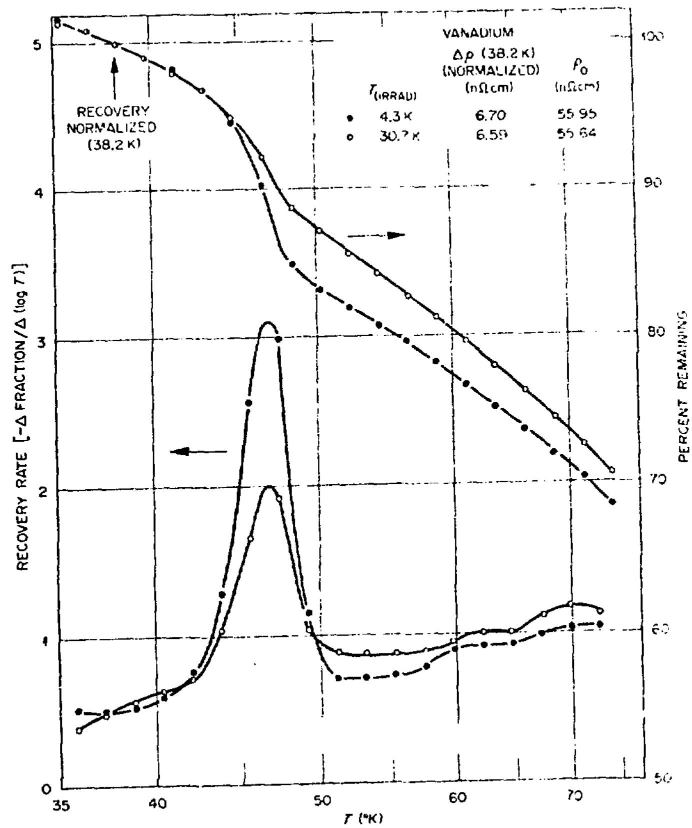

Fig. 2. Isochronal recovery and recovery rates after irradiation at 4.3 K and 30.7 K with equal doses of thermal neutrons.

ANALYSIS

Damage Rate.--Possible explanations for the decreasing damage rate at the low concentration involved in Fig. 1 are either (or both) (1) deviations from Matthiessen's rule, MRdev (on the additivity of different types of resistivity) or (2) partial recovery due to defect mobility and trapping at impurities during irradiation (Stage II production). Attempts to test these two explanations by an appropriate plot give a somewhat better fit to MRdev (where damage rate, $d\rho/dc$ is plotted vs $(c + c_o)^{-1}$; c is neutron capture event concentration and c_o is an adjustable constant) thar to Stage II production (where inverse damage rate, $dc/d\rho$ is plotted vs $\Delta\rho$, and, according to Wollenberger,[5] should be linear for impure samples). However, fits of this sort on data of this quality are not a very convincing basis on which to decide the mechanism. The similar behavior of the Pt case *must* be due to MRdev, since Stage I recovery does not begin until 10 K, let alone Stage II. So although these results favor MRdev for the V they do not rule out either mechanism.

47 K Peak.--The peak's sharpness (7.9% $\Delta T/\bar{T}$ full width at half maximum, compared to a theoretical width of 7.3% for a 1st-order process with the 4% $\Delta T/T$ isochronal schedule used) and smoothness call for a full analysis of the data to extract the activation energy.* The results in Fig. 3 show a remarkably extensive range of good fit which is strong evidence that the analysis is valid and that we do indeed have a pure first-order process with a single activation energy of 0.13 eV.

DISCUSSION

The observed sensitivity of the 47 K recovery to dose and to irradiation temperature rules out two of the possible conventional explanations: (1) a Stage I-A,B,C or D peak should be *in*sensitive to either of these variables and (2) an impurity detrapping process in Stage II should have a *negative*, not positive, dose dependence.

The properties of the 47 K peak seem to require a defect configuration which is not intrinsic to the irradiation but which is produced by migration of defects over long enough distances to undergo competing reactions with both other radiation defects and impurities. The resulting configuration might be either di-interstitials which break up at 47 K (and quickly

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*Combining the definitions of isochronal annealing and of first-order recovery yields an expression in which the ratio of fractions of the *process* defects remaining after successive anneals, (F_n/F_{n-1}), is related to the temperature, T_n; time, t; activation energy, E; and "frequency factor," α, as:

$$\log\,[-\log\,(F_n/F_{n-1})] = \log\,(\alpha t) - E/kT_n.$$

A plot of the quantity on the left vs $1/T_n$ should give a straight line from which E and α may be derived. In order to apply this analysis to real data, the process property change must be separated out from all other changes. In the present case, the peak is superimposed upon a continuous background which for this analysis was subtracted from the recovery data by using a linear interpolation between the background levels on either side of the peak.

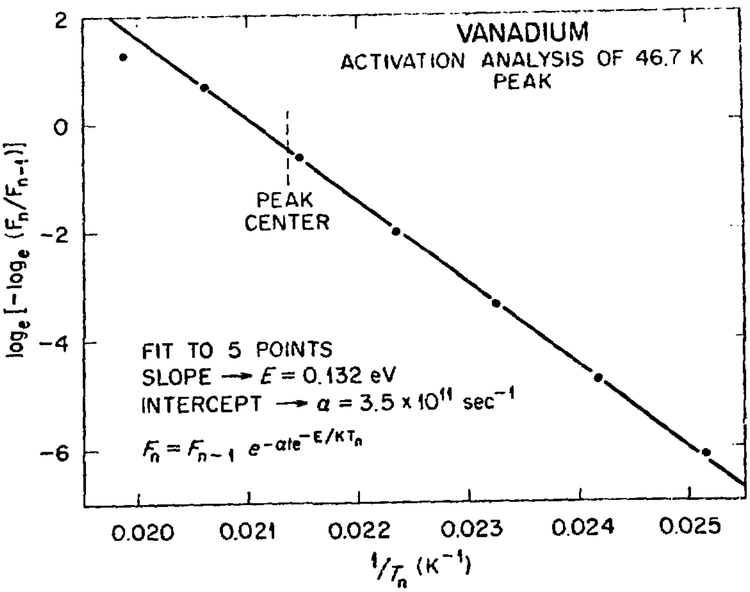

Fig. 3. Activation energy analysis of 46.7 K peak assuming a first-order process.

migrate to recovery) or a mutually trapped interstitial-vacancy pair which collapses at 47 K. The amount of recovery observed depends upon the population of this configuration which in turn is governed by the relative concentrations of defects and impurities prevailing during the generation process. These mechanisms, when combined with the decrease in impurity trapping with temperature implied by the 4 - 43 K continuous recovery, do yield the observed dose and irradiation temperature dependences. Explanation of the dose *in*dependence of that continuous recovery is not clear in this picture.

Further evidence suggestive of 4 K defect mobility is the low value of the Frenkel pair specific resistivity, $\rho_{FP} \simeq 6$ μΩcm/at.% derived by Chaplin[6] from electron irradiations of V at 6 K. Even though there may be other displacement probability functions than he used which would yield higher ρ_{FP}, this value is so very low compared to that predicted by the rough empirical rule[7] (1.5 x ρ_{273K}), namely 30 μΩcm, that there still remains considerable suspicion that defects are being lost during a 4 K irradiation.

Studies with purer material (which has been made[8] with RRR ~ 1800 in larger sizes than can be used here) and with very carefully controlled impurity additions and a wide range of doses are needed before we will really understand damage in V. If an ultra-low-temperature study were to show Stage 1, it would be definitive.

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